

Docket No.: 272909US0X PCT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:

GROUP: 1797

Elena BENCINI, et al.

SERIAL NO: 10/538,641

EXAMINER: SINGH, P.

FILED: November 9, 2005

FOR: CATALYTIC COMPOSITION AND PROCESS FOR THE
TRANSALKYLATION OF AROMATIC HYDROCARBONS

DECLARATION UNDER 37 C.F.R. § 1.132

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

Sir:

Now comes Elena Bencini who deposes and states that:

1. I am a graduate of Chemistry and received my degree in the year 1977.
2. I have been employed by Polimeri Europa since 2002 and I have been conducting research in the field of Catalysis for 22 years.
3. I am familiar with the prosecution history of the present application or the Office Action of April 7, 2010 was explained to me by counsel.
4. In order to demonstrate that catalytic compositions having a particular $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio and/or a particular fraction of pore volume having a diameter of greater than 100 nm provide substantially improved catalyst performance, the following experiments were carried out by me or under my direct supervision and control.
5. I have read and understood the Girotti (EP0847802); Grootjans (U.S. 5,750,814) and Ward (U.S. 4,185,040) patents cited by the Examiner; foreign equivalents thereof; or these references were explained to me by counsel.

6. Example 2 of the present application was repeated with a zeolite Y catalyst having a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio within the 10-20 range (i.e., a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 12). The first and second comparative examples of the present application were likewise repeated with zeolite Y materials having $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios outside the 10-20 range of the zeolite Y of the present claims. The comparative examples are identified as "CAT A-I" and "CAT B-I". The inventive and comparative catalysts are used to carry out a transalkylation reaction of polyethylbenzenes with benzene under the same conditions used in Example 2 of the present application. The results of the experiments are tabulated below.

	Zeolite Y $\text{SiO}_2/\text{Al}_2\text{O}_3$		Productivity (g EB/g ctz) 24	Productivity (g EB/g ctz) 145
Catalyst example 2	12	Polyethylbenzenes conversion %	80.1	78.9
CAT A-I	30	Polyethylbenzenes conversion %	70.1	69.3
CAT B-I	5.2	Polyethylbenzenes conversion %	80.4	72.1

7. The catalyst of inventive Example 2 and the catalyst of CAT A-I have the same porosity but are different in $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratios. The comparative catalyst provides a substantially lower conversion than the inventive catalyst; namely, a polyethylbenzenes conversion of 80.1% for the inventive example and 70.1% for the comparative example at a productivity 24 .

8. The catalyst of comparative Example CAT B-I may have an initial conversion similar to that of inventive Example 2; however, the comparative catalyst has a substantially greater decrease in polyethylbenzenes conversion over time. In the described range of productivity the catalyst of inventive Example 2 has a decrease in polyethylbenzenes conversion of only 1.2% absolute . Comparative catalyst CAT B-I has a corresponding reduction in polyethylbenzenes conversion of 8.3% absolute. It is readily evident that the

comparative catalyst having a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of less than 10 has a substantially shorter lifetime as evidenced by the decrease in polyethylbenzenes conversion over time.

9. The examples tabulated above and discussed in detail herein show that catalytic compositions containing the particular zeolite Y having a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio within the range of 10-20 are substantially superior to comparative catalytic compositions having a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of greater than 20, e.g., 30, with respect to the polyethylbenzenes conversion in the transalkylation reaction. The examples likewise show that a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio that is less than the minimum 10 required by Claim 1 is unable to provide the stability over time of the inventive catalytic zeolite Y.

10. Nothing in the Grootjans, Ward or Girotti references suggests that improved polyethylbenzenes conversion and catalyst lifetime can be obtained by using a zeolite Y having a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 10-20 in the manner recited in the present claims.

11. A further inventive example (Example A-II) was prepared and tested for transalkylation performance. The catalytic composition was based on a Y zeolite with a Si/Al ratio equal to 12 (CVB 712) and having the following characteristics:

Cat A-II = extrazeolitic pore volume: 0.85 cc/g

Porosity with diameter greater than 20 nm = 49%

Porosity with diameter greater than 100 nm = 34%

12. A catalytic test for the transalkylation of polyethylbenzenes with benzene was made using the same method in the same reactor used in Example 2 of the present invention and under the same operative conditions, i.e., 210°C reaction temperature, 50 bar reaction pressure, benzene/polyethylbenzenes molar ratio equal to 20, and a space velocity expressed as WHSV equal to 4h^{-1} .

13. After 45 hours on stream the product mixture was sampled and, based on a GC analysis, the polyethylbenzenes conversion was 92.6 %.

14. After 93 hours on stream the product mixture was sampled and, based on a GC analysis, the polyethylbenzenes conversion was 92.3 %.

15. A further comparative example (Example B-II) was prepared and tested for transalkylation performance. The catalytic composition was based on a Y zeolite with a Si/Al ratio equal to 15 (HSZ-630-HUA) and having the following characteristics:

Cat. B-II = extrazeolitic pore volume: 0.75 cc/g

Porosity with diameter greater than 20 nm: 52%

Porosity with diameter greater than 100 nm: 28%.

16. A catalytic test for the transalkylation of polyethylbenzenes with benzene was made using the same method in the same reactor used in Example 2 of the present invention and under the same operative conditions, i.e., 210°C reaction temperature, 50 bar reaction pressure, benzene/polyethylbenzenes molar ratio equal to 20, and a space velocity expressed as WHSV equal to 4h⁻¹.

17. After 24 hours on stream the product mixture was sampled and, based on a GC analysis, the polyethylbenzenes conversion was 22.6 %.

18. After 119 hours on stream the product mixture was sampled and, based on a GC analysis, the polyethylbenzenes conversion was 19.2 %.

19. The results of the experiments are tabulated below:

	Volume of pores with a diameter greater than 100 nm	Polyethylbenzenes conversion % (hours on stream)	
CAT A-II	28%	92.6% (45)	92.3% (93)
CAT B-II	34%	22.6% (24)	19.2% (119)

20. The comparisons described above show that the catalytic composition and process of the present claims are different from the catalytic composition and process described in the Girotti reference for at least the following reasons:

- the type of zeolite used (Y zeolite instead of beta zeolite)
- the porosity characteristic (at least 30% of pore volume consists of pores with a diameter greater than 100 nm),
- unexpectedly improved transalkylation of polyalkylaromatics.

21. The experiments described herein pertaining to catalytic compositions A-II and B-II show that for two catalytic compositions having substantially equal extrazeolitic volume porosity (i.e., 0.85 vs. 0.75 ml/g) and substantially equal porosity fraction with diameter greater than 20 nm, in percentage terms, (i.e., 49% vs. 52%), the inventive catalytic composition in which at least 30% of the total extrazeolitic volume has a diameter greater than 100 nm provides polyalkylaromatics conversion that is three times greater than polyalkylaromatics conversion provided by a comparative catalyst in which at least 30% of the total extrazeolitic volume does not have a diameter greater than 100 nm.

22. The catalytic composition B-II is based on a Y zeolite that is bound with alumina in a way that the porosity fully corresponds to the teaching of Girotti but does not have at least 30% of the total extrazeolitic volume having a diameter greater than 100 nm.

23. Nothing in the Grootjans, Ward or Girotti references suggests that improved polyaromatics conversion can be obtained by using a zeolite Y having a pore volume fraction of greater than 30% having a diameter of 100 nm.

24. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both,

under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

25. Further deponent saith not.

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Elina Bengim
Signature

4/2/2011
Date